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THE MINERALOGICAL MAGAZINE

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On the chemical classification of the mica group.

II. The basic micas.

By A. F. Hallimond, M.A., F.G.S.

Assistant Curator, Museum of Practical Geology, London.[1]

[Read January 19, 1926.]

In the first part of this paper[2] it was shown that the acid section of the mica group could be divided into two main series, the potash micas and the lithia micas. The molecular ratio of potash to silica was 1 : 6 and the micas were therefore represented as salts of an acid characterized by this value. The present section contains the result of a similar investigation of the basic micas, especially biotite and phlogopite ; here again it will be seen that the grouping $K_2O \cdot 6SiO_2$ is present throughout. For comparison of the molecular composition, the analyses of the basic micas have been recalculated in molecular ratios, $(Si,Ti)O_2$ being made equal to 600 according to the method already used for the acid micas. To economize space, however, figures have not been tabulated for the older analyses, but the values for R_2O_3 and RO, which are the only 'solid' variables when the ratio $K_2O \cdot 6SiO_2$ is fixed, have been

[1] Communicated by permission of the Director.

[2] A. F. Hallimond, Min. Mag., 1925, vol. 20, pp. 305-318.

plotted in fig. 5. This diagram renders it possible to ascertain at once those published analyses which approximate to a given composition; similarly, any new analysis can be classified and compared with earlier data. The analyses used in this paper are (1) the biotites and phlogopites cited by Dana,[1] (2) those given by H. E. Boeke (complete analyses later than 1890),[2] (3) a number of recent analyses by W. Kunitz and others; ratios for these are tabulated at the end of the paper.

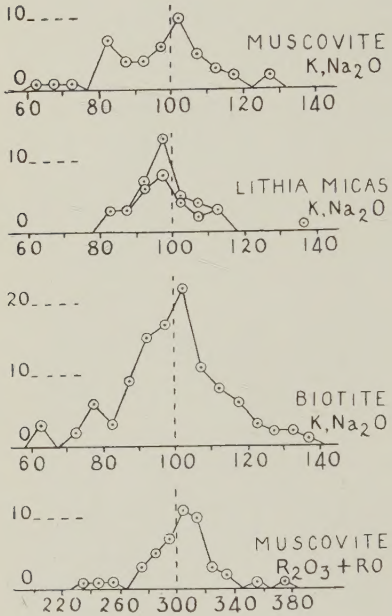


FIG. 3.[4] Frequency diagrams showing the incidence of R_2O-values in the published analyses of muscovite, lithia micas, and basic micas; also the incidence of values for $RO + R_2O_3$ in muscovite ($SiO_2 = 600$). The upper line for lithia micas includes a few analyses not listed in Part I. The ordinates represent the number of analyses occurring in each division of the base-line.

Ratio of SiO_2 to K_2O.—While the first part of this paper was in the press a discussion of the mica group was published by A. N. Winchell,[3] who assigns formulae with the ratios $K_2O \cdot 6SiO_2$ (phlogopite), $K_2O \cdot 5SiO_2$ ('eastonite'), and $K_2O \cdot 8SiO_2$ ('phengite'). It is of special interest, therefore, to examine what proportion of the mica analyses deviate from the ratio $K_2O \cdot 6SiO_2$. This is conveniently done by means of frequency diagrams (fig. 3), in which the base-line is divided into compartments, corresponding with intervals of, say, 60–65, 65–70, 70–75, &c., in the value for R_2O, when $SiO_2 = 600$. Above each compartment an ordinate is drawn representing the number of analyses for which the R_2O-value falls within the interval. The resulting points yield a curve showing the relative frequency of analyses for the respective values of R_2O. Three

[1] Dana's 'System of Mineralogy', 6th ed., 1892, pp. 630, 631, 633.

[2] H. E. Boeke, Neues Jahrb. Min., 1916, vol. 1, pp. 89–95. [Min. Abstr., vol. 1, p. 245.]

[3] A. N. Winchell, Amer. Journ. Sci., 1925, ser. 5, vol. 9, pp. 309–327, 415–450, diagram p. 321. [Min. Abstr., vol. 3, p. 12.]

[4] Figs. 1-2 were given in Part I of this paper.

such curves are shown in the figure, the first two being for the potash and lithia micas given in the first part of this paper, while the third represents the basic micas now plotted. The ratio $K_2O \cdot 5SiO_2$ requires the value 120 R_2O, while $K_2O \cdot 8SiO_2$ requires 75, and it is at once clear that few analyses contain R_2O in these proportions; the majority lie close to the value 100 and are symmetrically grouped in a manner which suggests that the deviations are due to accidental causes. The fourth curve in fig. 3 shows a similar distribution of the values for $R_2O_3 + RO$ in muscovite; these were shown in Part I to approximate to the simple value 300. Taken together, the four curves offer very strong

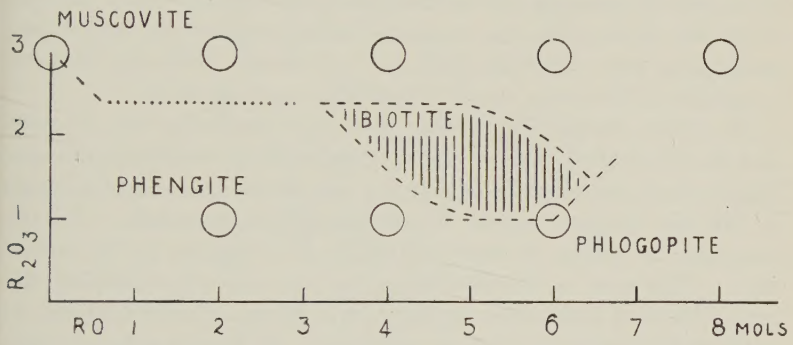


FIG. 4. Relation of acid to basic micas. Type $K_2O \cdot mRO \cdot nR_2O_3 \cdot 6SiO_2 \cdot 2H_2O$. General diagram showing the limits of composition for muscovite and biotite; circles indicate the compounds derivable from graphical formulae based on trisilicic acid.

support to the view that both acid and basic micas are essentially compounds of the constant group $K_2O \cdot 6SiO_2$.

Relation of muscovite to biotite.—Fig. 1 in Part I of this paper showed the composition of the muscovite group, the co-ordinates being the molecular proportions of RO and R_2O_3 when $SiO_2 = 600$. Fig. 5 is a similar diagram for the biotites. The two figures represent on a large scale portions of a general diagram for the muscovite-biotite group. This general diagram is shown in fig. 4, which summarizes the chemical relations of the two species. Only five analyses, tabulated on p. 33, fall in the space between biotite and muscovite, so that there is under normal conditions a wide gap in the range of solid solution.

Basicity of the micas in relation to that of the parent rocks.—The micas, with their widely varied range of RO-content, have presumably formed from silicate melts or solutions with a corresponding range of

composition. Beginning with melts of an acid character (pegmatites) nearly free from RO, we have, first, pure muscovite passing over into phengitic muscovite as the content of RO increases. In this way nearly one-third of the alumina may be replaced by RO. Further increase in RO results in the discontinuous addition of a substantial amount of RO to form the most acid members of the biotite group. 'Phase-rule' considerations would require that these acid biotites should be capable of co-existence with the corresponding muscovite, and they are indeed derived almost without exception from the granites and gneisses, rocks in which the two minerals often crystallize side by side. The range of micas from the granites and intermediate rocks extends about half-way across the biotite area, the remainder being occupied by vein micas, phlogopites, and a few micas from the basic rocks. Thus the chemical composition of the micas closely reflects their mode of origin.

The biotite group.—Fig. 5 represents very completely the chemical data for this section. Out of 119 analyses of biotite and phlogopite, only the five above mentioned lie outside the area of the figure and six others (p. 33) are omitted because of exceptional alkali-content. All the remainder lie within or very close to the area bounded by the broken lines. This area is therefore taken to represent approximately the normal limits of composition for the biotite group. Recent analyses are scattered fairly uniformly over the whole area, but there is need to supplement the existing data toward the top right and the bottom left corners. The latter contains three very interesting micas (nos. 201–203) from minette, kersantite, and syenite-porphyry, rocks which might be expected to yield biotites of exceptional composition. A rather sparse interval between biotite and phlogopite has now been bridged by recent analyses, so that the whole group seems to be a continuous series as regards composition. Micas usually called phlogopite comprise not only minerals of the accepted formula, but also a number represented by points toward the right of the area, with higher content of R_2O_3. 'Anomite' occurs in the centre of the area as well as on the phlogopite side, but a distinction based on the passage of 2V through zero does not seem to warrant making this a separate species. Lepidomelane, a term sometimes applied to dark ferrous micas, but usually to micas rich in ferric oxide, must certainly include a number of oxidized materials. The composition is well exhibited in the tables given by Doelter. On account of the frequency of alteration, the lepidomelanes given by Dana have not been plotted, but there can be no doubt that a few oxidized micas will have been included in fig. 5. Kunitz suggests that in most analyses some of the

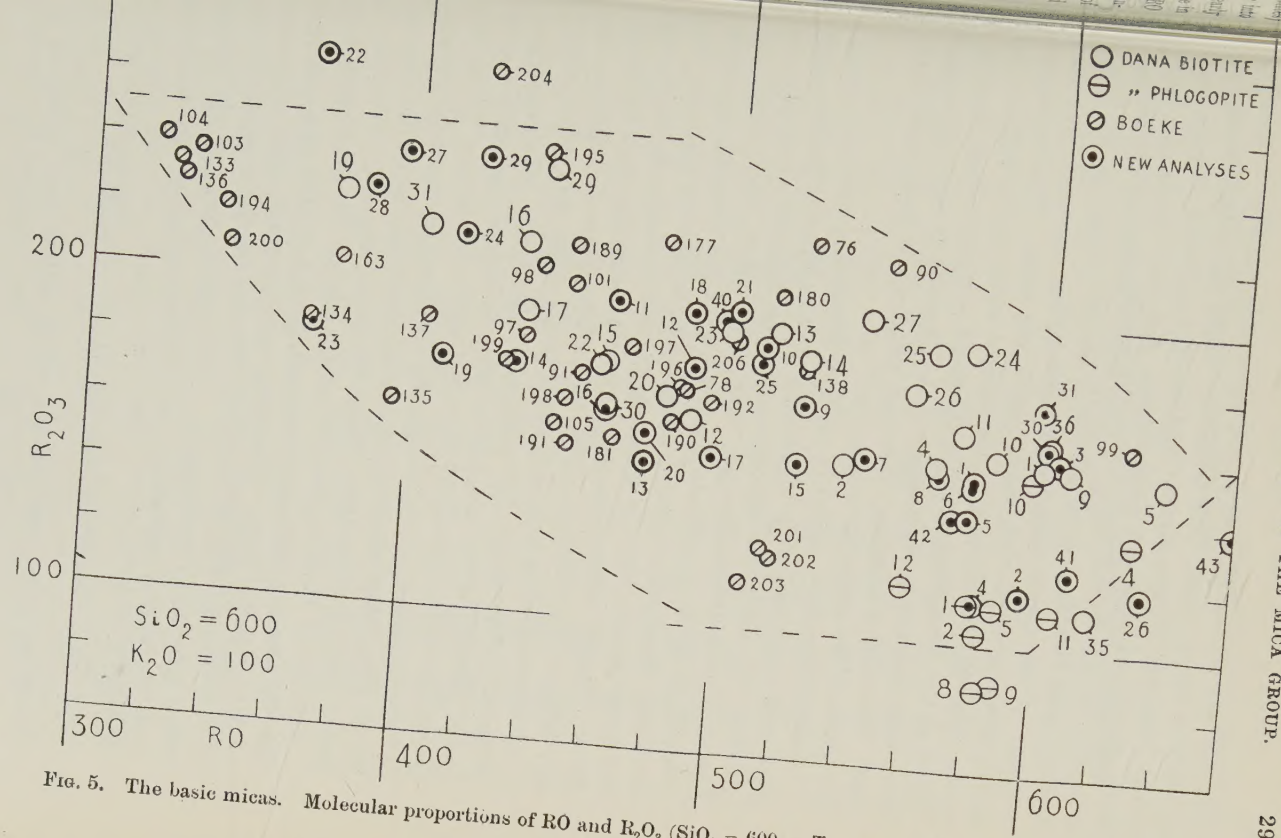
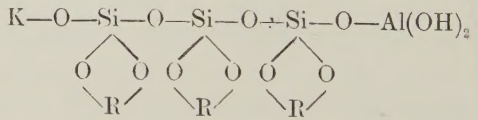


FIG. 5. The basic micas. Molecular proportions of RO and R_2O_3 ($SiO_2 = 600$). Type $K_2O \,.\, mRO \,.\, nR_2O_3 \,.\, 6SiO_2 \,.\, 2H_2O$.

water has been reduced by FeO during ignition, so that the values for H_2O tend to be too low. This would not affect the determination of ferrous iron, but Kunitz further suggests that a similar reaction has taken placein the mica after crystallization, and that all the iron was originally in the ferrous state. If the small amounts of ferric iron shown in most biotites were transferred to the RO group, the left-hand boundary of the biotite area would be somewhat narrowed, but the general character of the area would remain the same. There seems no reason, however, to believe that the natural melts are so deficient in oxygen as this assumption would imply, and it is more probable that the average fresh biotite is substantially in the condition in which it crystallized. The state of oxidation of iron and titanium will be discussed later in relation to the molecular volumes.

Chemical formulae.—In Part I the acid micas were referred to hexasilicic acid on account of the symmetry and convenience of the structural formulae. Similarly, the basic micas could be regarded as mixtures of a series of compounds of the type $K_2O \,.\, mRO \,.\, nR_2O_3 \,.\, 6SiO_2 \,.\, 2H_2O$, as indicated along the co-ordinate axes of fig. 4. Upon consideration of the most basic phlogopites, however, there appear reasons for preferring the half-formulae with KSi_3 as the common nucleus, similar to that given below. Possible trisilicic compounds are represented in fig. 4 by circles. According to the trisilicate scheme, the basic micas are essentially a series of mixtures extending from phlogopite, $\frac{1}{2}(K_2O \,.\, 6RO \,.\, R_2O_3 \,.\, 6SiO_2 \,.\, 2H_2O)$ towards a biotite $\frac{1}{2}(K_2O \,.\, 4RO \,.\, 3R_2O_3 \,.\, 6SiO_2 \,.\, 2H_2O)$. The series is extended laterally by the presence of other analogous compounds and is terminated at the acid end by the transition to muscovite. As the RO increases the series as a whole exhibits an increasing degree of substitution of R_2O_3 accompanied by an increasing addition of RO, so that the points extend down a rather oblique line on the diagram.

Biotites are easily represented graphically by a hexasilicate formula like that already given for protolithionite (Part I, p. 315), putting alumina or RO in place of lithia. Phlogopite, however, could only be represented by substituting RO for the water attached to the alumina, and this water is certainly not displaced. With trisilicic formulae, on the other hand, the valency available for the bases is greater; phlogopite can then be written



and from this the most basic micas on the right of fig. 5 are derived by putting $2(AlO_2R)$ or a similar group in place of R.

Basic Micas. Analyses recalculated to molecular proportions when $(Si,Ti)O_2 = 600$.

Table III. Recent analyses.

No.			R_2O_3.	RO.	$(K,Na)_2O$.	H_2O.	F_2.	$H_2O + F_2$.	V (obs.)[1]	V (calc.)	Diff
1	Kunitz,[2]	1 ...	151	578	105	325	—	325	323	312	11
2	,,	2 ...	114	595	83	55	119	174	299	300	−1
3	,,	3 ...	158	605	100	120	65	185	313	319	−6
4	,,	4 ...	113	580	89	42	92	134	—	—	—
5	,,	5 ...	139	577	103	137	52	189	310	307	3
6	,,	6 ...	149	578	114	162	42	204	316	311	5
7	,,	7 ...	156	544	108	175	—	—	—	—	—
8	,,	8 ...	152	567	100	152	44	196	309	311	−2
9	,,	9 ...	171	524	109	167	—	—	309	312	−3
10	,,	10 ...	188	511	110	171	4	175	319	317	2
11	,,	11 ...	199	464	99	217	—	—	315	314	1
12	,,	12 ...	180	489	101	239	—	—	314	310	4
13	Grout,[3]	1 ...	150	475	94	62	—	—	300	296	4
14	,,	2 ...	178	433	63	160	—	—	298	301	−3
15	,,	3 ...	153	523	89	59	—	—	300	304	−4
16	,,	4 ...	226	462	73	67	—	—	301	300	1
17	Seidel,[4]	1 ...	153	496	133	228	—	—	334	300	34
18	,,	2 ...	197	488	90	199	—	—	320	317	3
19	,,	3 ...	178	410	117	161	—	—	311	298	13
20	,,	4 ...	159	475	128	187	—	—	333	300	33
21	,,	5 ...	198	502	98	237	—	—	324	320	4
22	,,	6 ...	268	368	132	253	—	—	359	327	32
23	Pilipenko[5]	...	185	369	100	190	—	—	305	294	11
24	Stanley[6]	...	216	415	95	126	19	145	303	313	−10

[1] These values, for the molecular volumes, will be dealt with in a later part of the paper. Tables I–II were given in Part I.

[2] W. Kunitz, Neues Jahrb. Min., 1924, Beilage-Band 50, p. 386. [Min. Abstr., vol. 2, p. 424.]

[3] F. F. Grout, Amer. Min., 1924, vol. 9, p. 161. [Min. Abstr., vol. 3, p. 53.] Potash seems to be replaced by lime in nos. 14, 15, and 16.

[4] P. Seidel, Diss. Univ. Zürich, 1906. Refs. 4–8 are cited in Zeits. Krist., 1923, vol. 57, pp. 416–421.

[5] P. P. Pilipenko, Izvestiya Univ. Tomsk, 1915, no. 63, pp. 553–554. [Min. Abstr., vol. 2, p. 109.]

[6] E. R. Stanley, Trans. Roy. Soc. South Australia, 1916, vol. 40, pp. 268–271. [Min. Abstr., vol. 1, p. 70.]

Table III (*continued*) :

No.		R_2O_3.	RO.	$(K,Na)_2O$.	H_2O.	F_2.	H_2O+F_2.	V (obs.).	V (calc.).	Diff.
25	Thomassen [7]	183	510	102	53	3	56	—	—	—
26	Eckermann [8]	118	633	110	100	73	173	311	307	4
27	Harwood [9] ...	240	394	97	251	18	269	—	—	—
28	,, [10] ...	229	386	93	212	19	231	—	—	—
29	,, [11] ...	240	421	106	195	25	220	—	—	—
30	Vincent [12] ...	162	601	104	52	68	120	326	320	6
31	Ginzburg [13] ...	174	599	96	(ign.) 6	—	—	—	—	—
32	Jakob, [14] 1 ...	151	658	101	291	—	—	332	324	8
33	,, 2 ...	134	650	96	186	—	—	327	316	11
34	,, 3 ...	124	594	101	254	—	—	320	304	16
35	,, 4 ...	182	587	104	252	—	—	328	326	2
36	,, 5 ...	121	600	101	189	—	—	330	302	28
		(180)	(481)						(309)	(21)
37	,, 6 ...	206	545	113	213	—	—	327	329	−2
38	,, 7 ...	104	670	98	262	—	—	314	305	9
		(119)	(639)						(309)	(5)
39	,, 8 ...	97	669	92	169	—	—	310	302	8
		(126)	(611)						(307)	(3)
40	Eckermann [15]	195	498	100	183	7	190	316	318	−2
42	,, ...	123	610	108	144	67	211	313	305	8
43	,, ...	139	572	78	99	56	155	295	307	−12
44	,, ...	139	660	82	117	78	195	332	318	14

[7] L. Thomassen, Vidensk. Schrift. I. Mat.-naturw. Kl., Kristiania, 1920–1921, no. 9, p. 295.

[8] N. Sahlbom, Geol. För. Förh. Stockholm, 1922, vol. 44, pp. 383–385. [Min. Abstr., vol. 1, p. 396.] The figures given are for the analysis completed by Eckermann (see ref. 15 below).

[9] H. F. Harwood, Min. Mag., 1923, vol. 20, p. 23.

[10] Biotite from Dartmoor granite ; unpublished analysis included by courtesy of Messrs. A. Brammall and H. F. Harwood.

[11] H. F. Harwood, Min. Mag., 1924, vol. 20, p. 203.

[12] Phlogopite from Twinnge, Burma; unpublished analysis included by courtesy of Mr. H. C. G. Vincent (analyst).

[13] I. I. Ginzburg, Min. Abstr., 1925, vol. 2, p. 466.

[14] J. Jakob, Zeits. Krist., 1925, vol. 61, p. 157. [Min. Abstr., vol. 2, p. 423.] Lithia and fluorine do not seem to have been estimated. Except in nos. 5, 7, 8, iron and manganese are assigned to sesquioxide, since this brings the total nearer to 100. The use of the total in this way does not appear reliable and the values given in brackets have been calculated to show the result of regarding the (Fe,Mn) as sesquioxide throughout. Most of these analyses lie on the extreme right in fig. 5, but they have not been plotted on account of this uncertainty.

[15] H. Eckermann, Tschermaks Min. Petr. Mitt., 1925, vol. 38, p. 281. [Min. Abstr., vol. 3, p. 81.] Small amounts of Li_2O are included with R_2O_3. Manganese

Table IV. Basic micas not plotted in fig. 5.

	R_2O_3.	RO.	$(K,Na)_2O$.	H_2O.	F_2.	$H_2O + F_2$.
1. *Micas with high alkalis.*						
Boeke, 193 ...	195	449	136	230	—	—
,, 205 ...	145	424	135	182	—	—
,, 207 ...	226	442	130	266	—	—
2. *Micas with low alkalis.*						
Boeke, 146 ...	218	508	51	153	—	—
,, 179 ...	235	515	61	272	—	—
,, 176 ...	242	370	64	148	—	—
3. *Micas intermediate between biotite and muscovite.*						
Dana, 18 ...	278	203	93	86	111	187
,, 21 ...	322	296	111	268	7	275
,, 32 ...	303	255	97	265	—	—
Boeke, 80 ...	213	228	125	121	—	—
,, 100 ...	145	280	92	136	—	—

is not stated, and it seems possible that small amounts of that element may have been included with MgO. No. 43 contains also CaO (30 units) and BaO (7 units); the crystals were not transparent in bulk. See also no. 26, ref. 8, above.

Some mineralogical transformations in crystalline schists.

By C. E. Tilley, Ph.D., B.Sc., F.G.S.

Sedgwick Museum, Cambridge.

[Read January 19, 1926.]

THE twofold division of metamorphic rock-series into (*a*) Isophysical rock-series and (*b*) Isochemical rock-series, as proposed by P. Niggli[1] in his paper, 'Die Gesteinsassoziationen und ihre Entstehung', introduces into metamorphic petrology two very useful terms, which will be of service in the metamorphic researches that await the attention of the petrologist.

Among such investigated rock-series one may call to mind the studies on the chloritoid-schists of the St. Gotthard massif,[2] the glaucophane-schist suite of the Bagne valley (Wallis),[3] the calc-silicate series of the Trondhjem region,[4] the contact rocks of the Kristiania[5] and Comrie[6] areas. Of these the last are particularly illustrative of an isophysical rock-series, wherein a chemically wide group of assemblages conforming to the composition of shale-limestone sediments is represented in the free-silica and quartzless hornfelses. The crystalline schists of the Ordovician volcanic horizons of the Trondhjem region, studied by Carstens,[7] exemplify another isophysical series in dynamic metamorphism.

Among isochemical rock-series we turn to the pelitic crystalline

[1] P. Niggli, Verhandl. Schweiz. Naturf. Gesell., 1921, 101 Jahresvers. for 1920 in Neuenburg, part 2, pp. 123–147. [Min. Abstr., vol. 2, p. 315.] These terms are further defined and described in U. Grubenmann and P. Niggli, 'Die Gesteinsmetamorphose,' 1924, part I, pp. 481–484. Isophysical is synonymous with isograde or isofaciel used in conjunction with isograd, in treating metamorphism from the standpoint of the facies classification (Geol. Mag., 1924, vol. 61, p. 167).

[2] P. Niggli, Beitr. geol. Karte der Schweiz, 1912, N. Folge, Lief. 36, p. 1.

[3] T. J. Woyno, Neues Jahrb. Min., 1911, Beilage-Band 33, p. 136.

[4] V. M. Goldschmidt, Vid. Selsk. Skrifter, Kristiania, 1915, no. 10.

[5] Ibid., 1911, no. 1.

[6] C. E. Tilley, Quart. Journ. Geol. Soc. London, 1924, vol. 80, p. 22.

[7] C. W. Carstens, Norsk Geol. Tidsskr., 1922, vol. 7, p. 186.

schists of the Stavanger area [1] as the most thoroughly investigated, where detailed study by V. M. Goldschmidt reveals the chemical identity of varying mineral assemblages zonally arranged around Caledonian eruptives. Identical mineral assemblages were long ago recognized by G. Barrow [2] in his work on the crystalline schists of the south-east Highlands, but little detailed chemical work had been carried out.

The study of progressive metamorphism, whether it be concerned with the regional metamorphism of large mountain tracts or with the local metamorphism of small areolas surrounding intrusive rocks, brings constantly before the student two prime problems of determining,

(*a*) The genetic relations of the mineral assemblages of an isochemical rock-series,

(*b*) The varied mineral assemblages constituting particular isophysical rock-series.

Viewed from the standpoint of the facies classification these two problems resolve themselves into a study of the genesis of the critical index-minerals of the facies in an isochemical rock-series, and the broader problem of determining the mineral groupings of any one facies throughout the wide range of chemical composition revealed in rock masses. As I have expressed it elsewhere,[3] the elucidation of equivalent grades of metamorphism in a series of mixed sediments (or igneous rocks)—in a word, the determination of *isograde assemblages.*

It is hardly necessary to add that study along these lines is yet in its infancy. We are, so to speak, only at the threshold of an unexplored region, nevertheless the fruitful results already acquired in the pioneer studies of regional tracts, as the Scottish Highlands and the Scandinavian mountain area, lead one to hope that from such model metamorphic regions the solution to these problems will eventually be wrested.

In the realm of thermal metamorphism our knowledge is less scanty than in the case of regional metamorphism. This of course is due to two factors: firstly, more detailed investigations of thermal aureoles, and secondly, the data acquired in the laboratory investigation of silicate melts, have thrown a flood of light on the mineral relations obtaining in the inner zones of thermal aureoles. Much work yet remains to be done with the outer zones. Study here is intimately bound up with a determination of the constitution of very fine-grained sediments such as muds and shales. In the near future the application of X-ray studies to such

[1] V. M. Goldschmidt, Vid. Selsk. Skrifter, Kristiania, 1920, no. 10.
[2] G. Barrow, Quart. Journ. Geol. Soc. London, 1893, vol. 49, p. 330.
[3] C. E. Tilley, Geol. Mag., 1924, vol. 61, p. 167.

fine-grained material may lead to important results, and throw light on this vexed problem.

When the dynamically metamorphosed rocks come to be considered, we enter a region as yet unexplored by experimental research, a state of affairs brought painfully to our notice by the large number of unsynthesized minerals characteristic of crystalline schists. Progress has therefore been much slower in this branch of metamorphism; meanwhile an intensive field and laboratory study of crystalline schists in the light of physico-chemical data already accumulated should mark a great forward step. Those large mountain tracts particularly in which detailed mapping has revealed a great unbroken sequence of rocks in graded stages of metamorphism offer a wide and fruitful field for the metamorphic petrologist who may begin anew a quantitative study of the mineral assemblages and their spatial distribution. The first obvious line of attack is the elucidation of the varied mineral groupings in distinct isochemical rock-series that the particular metamorphic province has developed. This, indeed, was the method initiated by Barrow in his pioneer work of 1893, studies made more comprehensive by extended research resulting in the publication of the metamorphic map of the south-east Highlands in 1912.[1]

The pelite isochemical rock-series.—The dominant sediment type of such metamorphic provinces is the normal pelite, and it is therefore natural that our knowledge of the mineral assemblages of this chemical type is less incomplete than with other rock-series. Illustrative of such an isochemical series, it will serve to compare analyses of several mineral assemblages of the series, and these are quoted from the text-books in the accompanying table.

	I.	II.	III.	IV.	V.	VI.
SiO_2 ..	60·24	58·32	57·29	60·62	58·59	63·41
Al_2O_3 ...	18·46	20·00	20·60	18·50	19·48	19·22
Fe_2O_3 ...	2·56	2·01	2·40	1·32	1·07	2·74
FeO ...	5·18	4·98	5·48	5·65	5·31	4·00
MgO ...	2·33	1·85	1·76	2·42	3·31	2·84
CaO ...	0·33	0·66	0·37	1·66	1·72	0·65
Na_2O ...	1·57	1·26	1·40	1·81	1·65	1·21
K_2O ...	4·09	4·49	4·30	4·02	4·59	4·36
H_2O+ ...	3·99	4·10	4·05	3·09	3·24	0·60
TiO_2 ...	0·92	0·98	0·97	0·80	0·92	0·65
P_2O_5 ...	0·11	0·15	0·14	0·08	0·15	—
MnO ...	0·07	0·22	0·41	0·07	0·13	0·05
CO_2 ...	0·08	0·43	0·05	—	—	—
Accessories ...	0·22	0·57	1·02	0·05	—	—
	100·15	100·02	100·24	100·09	100·16	99·73

[1] G. Barrow, Proc. Geol. Assoc. London, 1912, vol. 23, p. 268.

I. Slate: Vermont, U.S.A.
II. Chlorite-muscovite-phyllite: Stavanger, Norway.
III. Spessartine-almandine-chlorite-mica-schist: Stavanger, Norway.
IV. Almandine-biotite-mica-schist: Stavanger, Norway.
V. Kyanite-staurolite-almandine-mica-schist: Ta-ku Shan, Liautang, China.
VI. Sillimanite-almandine-orthoclase-cordierite-gneiss: Södermanland, Sweden.

Despite the knowledge so far obtained of the various facies in this pelitic rock-series, little study has been made of the genetic relations obtaining among the constituent facies. Each mineral grouping has been studied rather as a complete assemblage, but a closer acquaintance with the mineralogical transformations that result in the passage from a lower to a higher assemblage is highly desirable. Here it is that the fuller recognition of the element of progression in metamorphism brings us into closer contact with the genetic aspect of the subject. To illustrate the point at issue, little is known concerning the nature of the mineralogical reactions which culminate in the production of the critical index-minerals of the zones of the rock-series now under discussion; for example, the genesis of (*a*) biotite, (*b*) almandine or spessartine, (*c*) staurolite or kyanite, (*d*) sillimanite.

Analyses of metamorphic rocks.—As representing originally mechanical mixtures of the detritus of pre-existing rocks and their degradation products, metamorphosed sediments constitute accidental assemblages, whose chemistry, obeying no simple law of distribution, is widely variable from point to point through any rock unit. From the nature of the case it follows that chemical analyses of sediments and their metamorphic derivative have not the significance of igneous rock analyses, which in themselves in large measure portray the stage of descent of the magmatic melt of which they are the record. Sediment analyses simply record the bulk composition of discontinuously varying mineral aggregates. Much care is necessary in the selection of such material for analysis, and it is not improbable that the wide disagreement sometimes observed between a rock analysis and the published petrographical description of the analysed rock is to be traced to an inhomogeneity of the selected material.

One further point deserves comment. The great majority of the minerals characteristic of crystalline schists are generated independent of the silica content of the rock over wide limits. As striking examples we need only mention the silica-poor, alumina-rich minerals of the chloritoid-ottrelite group, and also staurolite. Schists with chloritoid as a prominent constituent show silica percentages ranging in the wide

interval between 30 and 80. The passive role of silica must therefore be considered when analyses of sedimentary rocks are compared, and for the case of the pelitic isochemical rock-series the relations of a series of rock analyses are best discovered by a graphical plot of the oxide groups alumina, alkali oxides, and combined magnesia and ferrous oxide (all iron as FeO).

Analyses of metamorphic minerals.—When we turn to the available record of metamorphic minerals, we find a great gap in our knowledge of the composition of those minerals which are found widely distributed in the mass of crystalline schists. I refer now not to such minerals as cordierite or the scapolites, or stress minerals entirely confined to metamorphic rocks, but to those minerals common to both igneous rocks and metamorphosed sediments, particularly to the common micas, muscovite and biotite, pyroxenes and amphiboles, chlorites, and serpentines. Inspection of suites of analyses of the micas and the chlorites shows that the material chosen has come almost exclusively from igneous rocks, vein material, or well-crystallized material in druses and vughs. Analyses of these minerals extracted directly from the groundmass of slates, contact rocks, or crystalline schists are few indeed.

In the present unsatisfactory state of the knowledge of the constitution of these minerals it is obviously of great importance that such analyses should be carried out. Much attention has been devoted of late to the solution of the muscovite-biotite problem, and various complicated hypothetical formulae have been devised to explain the constitution of these micas, but it is clearly unprofitable for the metamorphic petrologist to substitute hypotheses for facts. The wide range of composition revealed in rock-making micas makes it imperative that analyses of the material in the setting of the crystalline schists should be available in order to understand the genetic problems connected with their metamorphism.

Graphical representation of analyses.—In order to visualize the analyses of mineral assemblages in the pelite isochemical rock-series, a triangular plot with alkalis, $FeO + MgO$, and Al_2O_3 as the three variables may be used with advantage. In order to utilize a greater area of the triangle, the three variables are taken as $K_2Al_2O_4$, $MgO + FeO$, and Al_2O_3; all Fe_2O_3 being converted to and treated as FeO.

Compositions of the muscovite-chlorite system.—In the accompanying diagram (fig. 1) the field *KMS* represents the composition field of mixtures of kaolin (or other hydrated aluminium silicate), muscovite ($H_2KAl_3Si_3O_{12}$), and the chlorites with iron-ores. This indeed should be the field of normal pelitic sediment as represented by shales and slates.

It is easy to test this by plotting in the recast analyses of slates. For this purpose thirty-two first-class analyses of slates have been taken free or almost free from CaO. The concentration field of these is also shown in fig. 1. It is seen that only half the field is enclosed within the area *KMS*. The extension of the field below *MS* is indicative either of a considerable percentage of orthoclase or albite in the slates, or the composition of the muscovites of slates does not correspond to the usual formula given by Tschermak. Most of the slates have been petrographi-

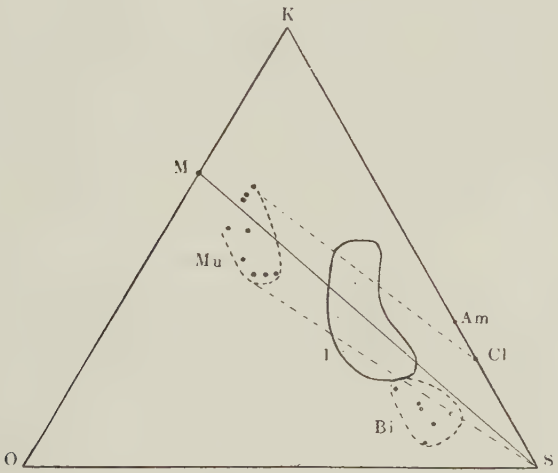


Fig. 1. Concentration field of slates (area 1).

[K = kaolin, O = orthoclase and albite, S = serpentine and iron-ores, Am = amesite, M = theoretical muscovite, Mu = field of muscovite from slates and mica-schists, Bi = field of biotite from mica-schists, Cl = limit of field of rock-making chlorites in sediments.]

cally described, and the published descriptions do not allude to the frequency of felspar, such as is demanded by the plot, and although fine-grained albite is likely to be mistaken for quartz, it may be doubted whether the assumed presence of felspar is the chief contributory cause of the discrepancy. These doubts are further strengthened when the few actual reliable analyses[1] of rock-making muscovites of slates and

[1] Quart. Journ. Geol. Soc. London, 1893, vol. 49, p. 355; Bull. U.S. Geol. Survey, 1910, no. 419, p. 286; Neues Jahrb. Min., 1912, vol. 1, p. 399; H. Rosenbusch, 'Elemente der Gesteinslehre,' 4th edit. by A. Osann, 1922–1923, p. 587; C. Doelter, 'Handbuch der Mineralchemie,' 1915, vol. 2, part 2, p. 422.

mica-schists are plotted in the diagram. These are plotted in the figure, and it is seen that the majority of them fall below the line *MS*.

In the case of chlorite, the scarcity of analyses of material present in the groundmass of slates or phyllites is even more apparent; but for our immediate purpose this desideratum is not so pressing, as the combination of chlorite with the accessory iron-ores is represented in the plot over a sufficiently wide range to cover great variation in the composition of the rock-making chlorites. The point *Cl* represents, in fact, the limit reached towards amesite in the available analyses of rock-making chlorites of pelites. The extended field revealed by the plotted positions of the muscovite analyses is now seen to cover a large part of the slate concentration field. The extension of the latter towards alumina indicates the presence of kaolin or related minerals, and doubtless the presence of detrital albite throws the field in the direction of *O*, but the few analyses represented in this part of the field do not give decisive evidence on this point by the relative potash-soda ratios.

Genesis of biotite.—As is well known, one of the first stages in the conversion of slates and phyllites into crystalline schists or into hornfelses on approaching igneous contacts is the production of biotite amidst the chlorite-sericite groundmass. We know, too, solely from optical determinations, that the composition of the biotite changes as metamorphism advances. Examination of any thermal aureole reveals that the little flakes of biotite as the igneous contact is approached gradually develop a deeper red tint, indicative, no doubt, of continuous enrichment in ferrous oxide. Similarly, in the case of the crystalline schists the earliest formed biotites are often green to brown-green in colour, eventually developing the common red-brown tint as the grade of metamorphism advances.[1] What other changes go hand in hand with this increase in FeO (if such it be) are quite uncertain. There does not appear to be recorded more than one analysis of biotite from pelitic hornfelses, that carried out by Jannasch in 1886. Better testimony of the limited knowledge of this aspect of the subject could scarcely be imagined.[2]

[1] C. E. Tilley, Quart. Journ. Geol. Soc. London, 1925, vol. 81, p. 103.

[2] A. F. Hallimond (Min. Mag., 1924, vol. 20, p. 193) and F. F. Grout and G. A. Thiel (Amer. Min., 1924, vol. 9, p. 228) have pointed out the possible importance of stilpnomelane as a constituent of metamorphosed sediments, especially in relation to its optical similarity to biotite. Lack of knowledge of the constitution of groundmass 'chlorites' and 'biotites' prevents as yet any estimate of the role accorded to stilpnomelane in metamorphosed sediments.

Boeke [1] in a noteworthy contribution to the muscovite-biotite problem has already clearly shown that Tschermak's conception of the constitution of the micas cannot be maintained, and this fact is clearly demonstrated in the plot of analyses [2] of rock-making biotites in pelitic rocks shown in fig. 1. After allowing for the inclusion of Fe_2O_3 as FeO, in the diagram, the analyses fall too far below the line *MS*.

The difficulty now arises that the composition field of biotite is not enclosed within the muscovite-chlorite field. That part of the slate belt below the line *MS* is, however, comprised within the muscovite-biotite field. Either the limited number of analyses of muscovite do not show the variation limit of rock-making muscovites, i.e. varieties still richer in potash exist, or else, as the fields muscovite-chlorite and muscovite-biotite have a common field, biotite is a product of a reaction between potash-rich muscovite and chlorite, with the accompanying formation of a muscovite less rich in potash. More analyses of the muscovites of slates and schists will be necessary before a solution of this problem is reached.

Muscovite-biotite-schists and almandine-biotite-schists.—In fig. 2 the concentration field of biotite- and biotite-garnet-schists, free from potash-felspar and aluminium silicates, in fact the rocks typical of the biotite and almandine zones, is plotted, based on twelve analyses. A large number of analyses of rocks of this type is not available. The field of these rocks is almost completely enclosed in the fields of muscovite-biotite-almandine (*Mu-Bi-A*). The field is much more restricted than in the case of slates. The reason for this is obvious. The metamorphic line of descent of slates is varied, some as will be shown later belong to the same isochemical series as the typical chloritoid- and staurolite-schists. As in the case of the slate field, there is an extension of this field below the lower limit of *Mu-Bi*, to be ascribed to the presence of albite.

Genesis of almandine.—The march of metamorphism of regional type in pelitic sediments leads on from muscovite-biotite-schists to garnet-bearing schists. The common garnet of the garnet zone is essentially an almandine usually with very low percentages of magnesia and ferric oxide. In manganese provinces, spessartine-almandine is readily

[1] H. E. Boeke, Neues Jahrb. Min., 1916, vol. 1, p. 83.

[2] Quart. Journ. Geol. Soc. London, 1893, vol. 49, p. 355; Nyt Mag. Naturv. 1886, vol. 30, p. 818; Mitt. Thurg. Naturf. Gesell., 1888, vol. 8, p. 13; H. Rosenbusch, 'Elemente der Gesteinslehre,' 4th edit. by A. Osann, 1922–1923, p. 655; Zeits. Krist., 1923, vol. 57, p. 416; C. Doelter, 'Handbuch der Mineralchemie,' 1916, vol. 2, part 2, p. 695.

generated, as analyses of the rocks of the phyllites of the Ardennes[1] first revealed. Here, indeed, the garnet arises at an earlier stage than the development of brown biotite. The presence of lime also probably favours early generation of garnet, and the grossular molecule is frequently not unimportant in analyses of almandines from pelitic rocks. The early generation of garnet in the Loch Lomond area, among the albite-schists, probably finds its explanation in the presence of lime as calcite in the pelitic sediment. The essential garnet molecule, however,

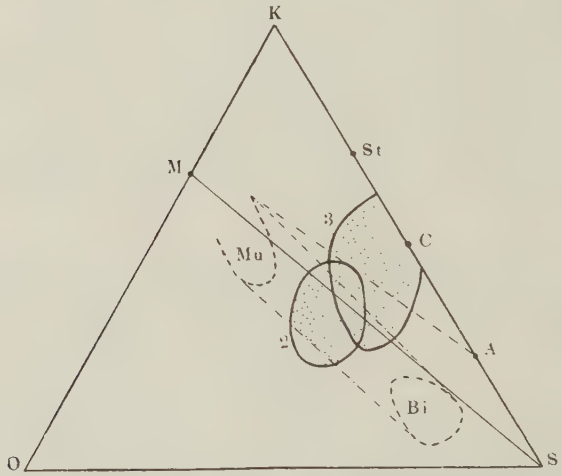


Fig. 2. Concentration field of mica- and garnet-mica-schists (2), and of chloritoid-schists (3).

[C = chloritoid, A = almandine, K = kyanite, St = staurolite.]

in metamorphosed normal pelitic sediment is almandine. Its genesis in the garnet zones of metamorphic provinces is still obscurely known. Two obvious sources suggest themselves, (*a*) chlorite, (*b*) biotite, to which may be added as contributor iron-ores.

The fact that it is possible to draw garnet isograds in regions of progressive metamorphism, points clearly to the conclusion that one dominant reaction is involved in the first production of almandine. Examination of those rocks situate at the first points of entry of garnet in pelitic sediments in the southern Highlands, frequently reveals the garnets in skeleton form, growing at the expense of the surrounding

[1] A. Renard, Bull. Mus Roy. Hist. Nat. Belg., 1882, vol. 1, p. 10.

minerals. In thin slices the development of such skeleton and amoeboid almandines from laths and scales of chlorite, interspersed among quartz grains, is shown by mica-schists occurring along the garnet isograd. The relations there seen make it quite clear that garnet is arising from chlorite. The reverse process of chlorite arising on the ruins of garnet, is common enough in petrographical observation, but the direct development of garnet from chlorite is much less familiar. To understand the

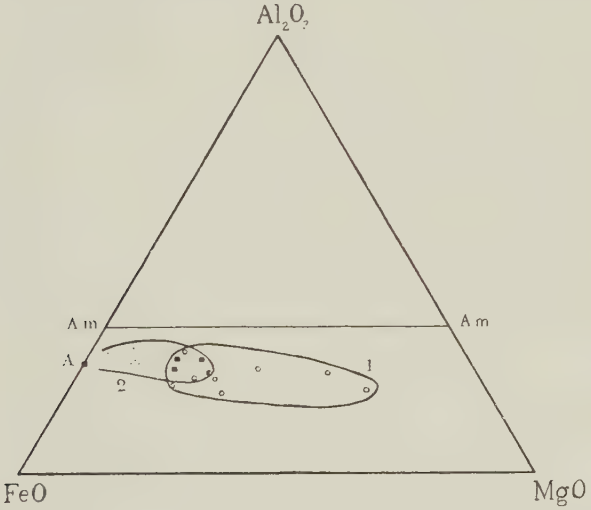


Fig. 3. Concentration fields of chlorite (1) and almandine (2) in pelites.
[A = almandine, Am = amesite end-members.]

mechanism of this production analyses of rock-forming chlorites and almandines in pelites must be reviewed.

As in the case of muscovite and biotite, there are few available analyses of rock-making chlorites in pelites. These [1] are plotted in fig. 3 (field 1), FeO being separated from MgO. The field of almandines of pelites is also indicated (field 2). Considering the ratio RO/Al_2O_3 it is seen that there is correspondence in the two minerals; were that the only consideration, the formation of garnet from chlorite would only

[1] C. Hintze, 'Handbuch der Mineralogie,' 1897, vol. 2, p. 728; M. F. Heddle, 'Mineralogy of Scotland,' 1901, vol. 2, p. 125; H. Rosenbusch, 'Elemente der Gesteinslehre,' 4th edit. by A. Osann, 1922–1923, p. 587; Neues Jahrb. Min., 1897, vol. 2, pp. 156, 215; Amer. Journ. Sci., 1886, ser. 3, vol. 32, p. 307.

require addition of silica. But it is in the ratio FeO/MgO that rock-making chlorites and almandines reveal discrepancies. The two fields overlap, this overlap being based on four analyses[1] of garnets (Fort Wrangel, Wittichen, Dachsbusch, Laacher See).

Almandines preponderate in the left-hand part of the field, not overlapped by the chlorite field. Whilst then some chlorites have the appropriate FeO/MgO ratio of that of actual almandines, many almandines cannot be thus satisfied. Nor can they be satisfied if it be assumed that iron-ores take part in the reaction, as will be seen from the diagram.

As specific examples of this non-agreement we may note the careful analyses of Penfield on the composition of almandine and the chlorite resulting from its degradation, for the case of the Lake Superior and Salida almandines, and further the compositions of the garnets and chlorites of the schists of the Stavanger region. In these cases the chlorites are all much richer in magnesia than the garnets. Actual exchange of magnesia for ferrous oxide, alumina being unchanged, is suggested. The position is still further complicated by the fact that almandines frequently contain appreciable percentages of the grossular and spessartine molecules, which cannot be accounted for in chlorite analyses.

One of the standard equations for the production of garnet from biotite is given in the reaction, biotite + quartz = almandine + orthoclase. There is a considerable body of evidence in support of such a development in high-grade pelites, but I believe this reaction is not typical of the almandine zone proper. In the garnet zone of the southern Highlands, orthoclase is a rare mineral, and where present can generally be assigned to its original presence as a detrital mineral (as in arkoses, cf. the Moine gneisses). It is, of course, to be understood that these remarks apply specifically to pelitic sediments. In the metamorphosed calcareous sediments of the almandine zone, potash-felspar may be abundantly generated, as the calc-silicate rocks of the Highlands testify. If garnet arises from biotite in the almandine zone, such a reaction finds no place, and a biotite derivation at this early stage has, I believe, yet to be demonstrated.

Chloritoid-rocks.—The concentration field of the chloritoid-schists, based on thirty analyses, is shown in fig. 2 (field 3). It is seen that chloritoid-schists cannot be strictly considered in the same isochemical series as the pelitic mica-schists, though the two fields overlap. The

[1] Dana, 'System of Mineralogy,' 1892, p. 442; C. Doelter, 'Handbuch der Mineralchemie,' 1921, vol. 2, part 3, pp. 365, 368.

principal distictions are lower alkalis, and in part higher alumina and iron oxides. The most complete treatment of the chloritoid-schists is that given by Niggli in the paper already quoted. Chloritoid begins already to form in the chlorite zone, at the expense of kaolin or sericite in the presence of iron oxides, and persists into higher zones.

Staurolite-rocks.—The concentration field of the staurolite-rocks is practically coincident with that of the chloritoid field of fig. 2, and these rocks are to be considered in one and the same isochemical series. Van Hise [1] has stated that 'staurolite rocks usually contain orthoclase or at least some potash felspar'. This statement I believe not to represent the facts. In the typical staurolite-rocks of the Highlands and the St. Gotthard region, potash-felspar is absent, nor is there evidence of its former presence, and it is to be doubted whether potash-felspar is generated in pelites in regional metamorphism below the metamorphic grade of the sillimanite zone. The generation of staurolite from chloritoid, of which evidence is provided in the staurolite-rocks of the Piz Scopi and other areas, is doubtless one of the principal reactions by which this mineral is developed in the crystalline schists. The evidence for this transformation I have already summarized in a previous paper.[2]

Index-minerals of pelites and the depth zones.—The index-minerals, biotite, almandine, staurolite, kyanite, in their pelitic assemblages are lumped together in the meso-zone of Grubenmann's classification of the crystalline schists. Chlorite is relegated to the epi-zone, and sillimanite allotted to the kata-zone. It is such examples as these that demonstrate the inadequacy of the depth zone hypothesis to express the diversity of mineral assemblage in an isochemical rock-series. It is to the credit of the facies classification that the finer expression of this mineral diversity is adequately provided for. The recognition of metamorphic zones, mapped with precision in regional tracts of crystalline schists, is clearly in harmony with the scheme of the facies classification, which, based on physical and chemical principles, yet freed from any concept of depth or *a priori* pigeon-holing, can grow in strength and completeness with the onward march of petrological research.

Conclusion.—If the foregoing remarks have settled little, they at least have demonstrated some of the desiderata to be acquired before any adequate conception is obtained of the chemical reactions taking place in what is probably the most intensively investigated of the isochemical rock-series. In view of the wide range of composition of the constituent

[1] C. R. Van Hise, Monograph U.S. Geol. Survey, 1904, no. 47, p. 328.

[2] C. E. Tilley, Geol. Mag., 1925, vol. 62, p. 318.

minerals, elaborate calculations of their composition, starting from the bulk analysis of the metamorphic rock, are to be looked upon with grave suspicion. The mechanism of the mineralogical transformations in crystalline schists is something much more complex than is explained away by the customary simple equations. The most pressing need is a closer chemical investigation of the common minerals occurring in the groundmass of metamorphic rocks.

On garnet in pelitic contact-zones.

By C. E. Tilley, B.Sc., Ph.D., F.G.S.

Sedgwick Museum, Cambridge.

[Read March 16, 1926.]

Of the garnet group of minerals, two members only appear to have been synthesized from artificial melts, viz. melanite (L. Michel, 1892) and spessartine (L. Bourgeois, 1883, and A. Gorgeu, 1883); though grossular is once reported by E. S. Shepherd, G. A. Rankin, and F. E. Wright (1909) as being produced by reaction under pressure of calcium orthosilicate and aluminium chloride. In dry melts, pyrope is represented by a cordierite-forsterite-spinel assemblage, but no data are yet available to indicate the dry melt assemblages representing almandine. The home of almandine in sediments is the dynamically metamorphosed pelite, and its widespread regional occurrence in these crystalline schists implies a ready synthesis under the influence of shearing stress.

While stress in this case appears to play the part of a catalyst, a garnet (customarily assumed as almandine) is also found in pelitic contact-rocks, though it is inconstant and sporadic in its occurrence. Examination of the thermal assemblages of a considerable number of contact-zones reveals that almandine is completely absent. Thus in the case of the Kristiania and Comrie contact-zones, though many hundreds of thin sections of the thermal products have been examined, no examples have been found to contain almandine. With one or two isolated exceptions, the same remark would appear to apply to the contact-zones of the Hercynian granites of Cornwall and Devon. In certain aureoles, however, garnet becomes a conspicuous and significant mineral.

To confine attention to the aureoles of the British Isles, we may note its occurrence in the contact-zones of:

(*a*) The Skiddaw granite and Grainsgill greisen; Carrock Fell gabbro.

(*b*) The New Galloway granites (Loch Dee and Cairnsmore of Fleet masses).

(*c*) Leinster granite and Glendalough amphibolites (Co. Wicklow).

(*d*) Insch and Huntly younger intrusives, Banffshire.

(*e*) Carn Chuinneag granite, Ross-shire.

The conditions of genesis in these aureoles have been hitherto unexplained and remain a puzzling petrographic problem.

Destruction of almandine in contact-zones of crystalline schists.—The absence of almandine in the typical contact-zones already mentioned is of such significance that an examination of aureoles of pelitic crystalline schists may throw some light on this question. It is unfortunate that our information in this direction is still limited; nevertheless some pertinent data are already at hand. A number of the Caledonian intrusions of the Scottish Highlands cut well-defined crystalline schists, of which almandine is a prominent constituent. It will suffice to mention the aureoles of the Cruachan, Ross of Mull, and Garabal Hill massifs. In each of these cases hornfelses have been manufactured out of garnet-bearing muscovite-biotite-schists, and in the three aureoles the destruction of almandine can be followed in the regeneration that has resulted. Particularly convincing is an example in the Ben Cruachan aureole where E. B. Bailey (1916) has described the development of hornfelses from garnet-bearing Leven schists, in which the garnet is in process of dissolution, being replaced by and converted into pseudomorphs of cordierite and magnetite, while white mica gives rise to abundant potash-felspar. The same destruction of almandine with the formation of cordierite in its place may be recognized in the other aureoles mentioned, those of the Ross of Mull and Garabal Hill. The mineralogical transformation here revealed is obviously of great importance as attesting the instability of almandine under the conditions which commonly prevail in contact-zones.

The foregoing facts make it obvious that a closer examination of the garnet-bearing aureoles already mentioned is demanded if we are to have insight into the conditions responsible for the production of garnet. It will first be necessary to determine that the garnet in question is essentially almandine; here it is that such information is not forthcoming, as there are no available analyses of the garnets of the Skiddaw, New Galloway, Glendalough, or Carn Chuinneag aureoles. In pelitic stress-zones it is now known that the garnets in some cases contain considerable percentages of the spessartine molecule, and the presence of significant percentages of MnO in the sediment has a profound influence on the order of entry of the index-minerals in progressive metamorphism, the common order chlorite–biotite–garnet being inverted to chlorite–garnet–biotite. With this knowledge before us, it is pertinent to inquire whether the garnets of pelitic contact-zones are characterized by any similar peculiarity.

As regards analytical data for British aureoles, we are practically limited to an analysis by W. J. Sollas[1] of a garnet-hornfels and its constituent minerals, from the Leinster granite contact, near Carrickmines, Co. Dublin. The analysis of the garnet shows it to be a spessartine-rich type, as follows: SiO_2 37·63, Al_2O_3 16·43, Fe_2O_3 7·85, FeO 14·59, MnO 18·55, CaO 3·49, MgO 2·10. This analysis may be expressed in atomic percentages as $Fe_{40}Mn_{40}Mg_{10}Ca_{10}$.

In order to obtain further information as to the composition of the contact garnets, two partial analyses have been carried out of (*a*) garnet from a garnet-sillimanite-contact-rock from Knocknairling Hill, New Galloway, and (*b*) garnet from a metamorphosed grit at Grainsgill (Cumberland). The New Galloway garnet yielded on analysis SiO_2 37·80, Al_2O_3 20·79, Fe_2O_3 0·43, FeO 21·87, MnO 14·88, CaO 1·60, MgO 1·75, which can be expressed as $Fe_{52}Mn_{36}Mg_7Ca_5$; while for the Grainsgill garnet analysis gave FeO 30·16, MnO 6·02, expressed as $Fe_{70}Mn_{14}$.

The most striking feature of the garnet-rocks of the aureole of the New Galloway granites is the abundance of white potash-mica, in which indeed the garnets are not infrequently enclosed. The other common associates in addition to quartz are sillimanite and biotite. Cordierite is much less common, while potash-felspar, sometimes wholly absent, appears to be never more than an accessory constituent. Quite similar features characterize the contact-metamorphosed Ordovician slates of Co. Wicklow (J. A. Thomson, 1908).

The sharpest contrast is found in the assemblages of the aureoles of Kristiania or Comrie. In these, garnet is absent, cordierite is dominant, potash-felspar is present to the exclusion of white mica, which here is never a primary mineral. When we turn to the garnet-bearing rocks of the Skiddaw area, the contrast is not so sharp, but is nevertheless marked. Throughout the Skiddaw aureole generally, potash-felspar is a subordinate constituent, while white mica as a primary mineral is common, as of course is also biotite. These conditions hold for the garnet-bearing types. Comparing the two types of assemblages, it may be considered that in their abundance of cordierite, potash-felspar, and magnetite to the exclusion of white mica, the Kristiania-Comrie aureoles were relatively dry, and that the garnet-bearing aureoles in their paucity of potash-felspar and abundant micas were relatively wet.

Turning to igneous rocks (Archaean tracts apart) we find that spessartine or spessartine-almandine is a characteristic mineral of certain granite-pegmatites, and even of druses and miaroles of hypabyssal and

[1] W. J. Sollas, Sci. Proc. R. Dublin Soc., 1891, n. ser., vol. 7, p. 49.

volcanic rocks, where its association with pneumatolytic minerals, as topaz, tourmaline, and beryl, betrays the presence of residual solutions rich in mineralizers. The absence of almandine in the normal contact-aureole, the visible evidence of its replacement when introduced into the sphere of influence of igneous intrusions, conspire to demonstrate that the iron-garnet is unstable under the conditions prevailing in normal contact-metamorphism. When the garnet of garnet-bearing aureoles is shown to contain notable quantities of MnO, and when further such garnets are inconstant and sporadic in their occurrence (suggesting local concentration of manganese), it would appear legitimate to conclude that the formation of garnet in pelite aureoles is conditioned by the presence of MnO in the sediment, leading to the formation of a spessartine-bearing type. This is the more acceptable when it is realized that spessartine is readily synthesized in artificial melts. It is as yet impossible to estimate what contributory part, if any, the presence of abundant water during metamorphism—as the principal garnet-bearing aureoles reveal—plays in producing a concentration of an orthosilicate molecule as garnet. The essence of these remarks is the stress laid upon the presence of MnO as promoting the formation of garnet in contact-zones of pelites.

Smithsonite from the Rhodesia Broken Hill mines.

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[Read March 16, 1926.]

DURING an examination of zinc ores I had occasion recently to employ methods for the identification of smithsonite, and was surprised to find how scanty and indefinite are the data of its physical properties as stated in text-books. Most of the smithsonite known occurs either compact or as very small crystals many of which are of a composite nature. In this connexion Dr. L. J. Spencer recently acquired for the British Museum some very fine crystals from the Rhodesia Broken Hill mines, and he suggested that this material would be suitable for a detailed examination. These consist of two specimens obtained in 1921 from Mr. W. E. Harris [B.M. 1921, 769] and in 1923 from Dr. O. L. Brady [B.M. 1923, 619] respectively, and are portions of the same mass. Smithsonite crystals were first described from this locality by Dr. Spencer in 1908,[1] but these crystals were minute and showed only the forms $v\,(20\bar{1})$ and $s\,(22\bar{3})$.

The crystals now described are all simple rhombohedra $f(1\bar{1}1)$ with edges up to 7 mm. in length. On damaged crystals these edges are frequently truncated symmetrically by cleavage. Some apparent faces proved to be merely surfaces of contact of aggregated crystals, and, although well-defined, gave no reflected images. All the rhombohedral faces bear striations parallel to their longer diagonal, whereas similar crystals [B.M. 1924, 1050] obtained more recently from the same locality have their faces distorted by parallel growth as is common in calcite.

[1] L. J. Spencer, Min. Mag., 1908, vol. 15, p. 35.

Since the faces are not rounded, this striation may be due to a composite growth of individuals relatively oriented by a slight rotation about the trigonal axis.

This composite character is borne out by reflections from the cleavage-surfaces. Only on minute cleavage-rhombs do the surfaces give single images; ten such fragments showing good images gave a cleavage-angle varying from 72° 19′ to 72° 25′ with an average of 72° 21′, whereas larger fragments gave readings of anything between 72° and 73°. For comparison, an attempt was made to measure the cleavage-angle of the more-recently obtained composite crystals, which, moreover, differ also in having a very faint yellowish tint. Several cleavage-rhombs gave good single-image reflections, the readings for the cleavage-angle varying from 72° 21½′ to 72° 27′ with an average of 72° 24′.

The hardness varies from 4 to 4½. Owing to the cleavage, it is difficult to test how far the mineral scratches fluorite, but it was always possible to scratch a cleavage-surface with apatite. The hardness of smithsonite was first determined by Robert Jameson, who in his 'System of Mineralogy' (3rd edition, 1820, vol. 2, p. 442) says 'It is as hard as apatite'. Thereafter the hardness of smithsonite has been quoted as 5.

The crystals are in general quite colourless and transparent, though some of them appear to be yellowish through admixture of limonite, with which they occur intimately associated. A little over a gram of pure material was collected and two independent determinations of the specific gravity made on 0·6785 gram with a pyknometer gave 4·397 and 4·398 at 16·2° C. and 16·6° C. respectively.

For the chemical analysis, the powdered material was divided into two portions. Preliminary tests showed the impurities to consist of FeO, MnO, CaO, MgO, and the powder after drying at 110°C. contained no water. Consequently the carbon dioxide was determined by ignition-loss in both portions. In the former ZnO, FeO, CaO, MgO were determined in the usual way, and in the latter MnO was determined colorimetrically.

On ignition the powder becomes orange-coloured (zinc oxide), and on cooling it retains a reddish-brown ferruginous tint. When the ignited material is dissolved in hydrochloric acid, ferric oxide is left behind and only dissolves very slowly or by the addition of nitric acid. The original carbonate dissolves completely and with ease in hydrochloric acid, showing that the ferric oxide is produced during ignition. Consequently, to the value of CO_2 obtained by ignition must be added the oxygen equivalent to the oxidation of the ferrous oxide.

Results of analysis.

	I.	II.	Average.		
ZnO	63·18	—	63·18	$ZnCO_3$ ...	97·34
FeO	0·58	—	0·58	$FeCO_3$ ...	0·93
MnO	—	0·01	0·01	$MnCO_3$ ...	0·02
CaO	0·27	—	0·27	$CaCO_3$ ...	0·48
MgO	0·45	—	0·45	$MgCO_3$ ...	0·94
CO_2	35·34	35·36	35·35		
H_2O (hygroscopic)	0·03	0·04	0·04		99·71
			99·88		

The optical properties were determined on two detached crystals previous to the analysis for which waste fragments were preserved. From one a 60°-prism (prism 1) was ground with its edge parallel to the trigonal axis of the crystal and its faces roughly parallel to $m(2\bar{1}\bar{1})$; the prism-angle was measured as 61° 9′. The other crystal was cleaved into two, and two prisms (prisms 2 and 3) were prepared with the edges perpendicular and parallel to the principal axis and with prism-angles of 60° 56′ and 57° 56′ respectively. In prism 2 one of the faces corresponded roughly to $f(1\bar{1}1)$, and the orientation of prism 3 was similar to that of prism 1. The grinding was done with a Thomas & Smith instrument on a glass plate, putty-powder being used for the final polishing.

The refractive indices were determined with these three prisms for different wave-lengths by employing the Tutton monochromatic illuminator, for the calibration of which I am indebted to Dr. Herbert Smith, and gave the following results:

	Extraordinary ray.			Ordinary ray.		
Line.	1.	2.	3.	1.	2.	3
Li ...	1·6187	1·6187	1·6186	1·8427	1·8424	1·8423
C ...	1·6190	1·6190	1·6189	1·8436	1·8427	1·8433
Na ...	1·6214	1·6214	1·6212	1·8488	1·8480	1·8485
Tl ...	1·6243	1·6237	1·6239	1·8550	1·8545	1·8547
F ...	1·6277	1·6270	1·6274	1·8623	—	1·8621
G ...	1·6327	1·6320	1·6326	1·8737	—	1·8736

With prism 2 the violet end of the spectrum gave very faint images, and in the case of the ordinary ray the light was obstructed by a flaw. The results of prism 3 are probably nearest the truth.

Optical values for smithsonite are very few in the literature, and all those in the text-books appear to be derived from the following two sources. W. Ortloff[1] gave $\epsilon = 1{\cdot}61766$ on analysed pure material from

[1] W. Ortloff, Zeits. physik. Chem., 1896, vol. 19, p. 215.

Aachen. Sir Henry A. Miers in his 'Mineralogy' (1902, p. 553) gave the birefringence as 0·200, but I have been unable to trace the origin of this figure. Until recently these figures were the only ones published, when P. Gaubert[1] gave $\epsilon = 1{\cdot}612$ and $\omega = 1{\cdot}872$ for somewhat impure ($ZnCO_3 = 90{\cdot}43\,\%$) analysed material from Chessy; he also gave $\epsilon = 1{\cdot}619$ for smithsonite from Laurion. The value of ω he had previously given[2] as $\omega = 1{\cdot}782$, and I understand from Professor A. N. Winchell that P. Gaubert ascribes the higher value ($\omega = 1{\cdot}872$) to a misprint; moreover these values for ω were determined only indirectly from the extreme values of the extraordinary refractive index given by total reflection from the cleavage-surfaces.

Attempts to determine the indices by this method gave for the Rhodesian material $\epsilon = 1{\cdot}620$ and $\epsilon' = 1{\cdot}740$, from which, assuming $cr = 42°\,57'$, $\omega = 1{\cdot}838$. The readings of the refractometer were, however, very obscure.

Summary of Data.

Crystal-form $f(1\bar{1}1)$. Cleavage-angle $rr' = 72°\,21'$. H = 4–4½. G = 4·398. $\epsilon_{Na} = 1{\cdot}6212$, $\omega_{Na} = 1{\cdot}8485$. Contains 97·34 % $ZnCO_3$.

[1] P. Gaubert, Bull. Soc. Franç. Min., 1919, vol. 42, p. 99.
[2] P. Gaubert, Compt. Rend. Acad. Sci. Paris, 1917, vol. 164, p. 47.

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